POTENTIAL OF NON-WOOD FIBRES FOR PULP AND PAPER-BASED INDUSTRIES

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ABSTRACT

The global demand for wood fibre has increased due to increasing population and new applications for wood fibre. Therefore, to supplement the limited wood fibre resources, non-wood fibres have been introduced as alternatives in pulp and paper-based industries. This study aims to use non-woods as pulp in paper-making industry: promoting the concept of "from waste to wealth" and "recyclable material". Hence, the objective of this study is to determine the potential of cassava peel, cocoa pod husk, cogon grass and oil palm leaf as alternative fibres for pulp in paper-based industries based on its chemical, physical and mechanical properties. The chemical properties involved in this study (holocellulose, cellulose, hemicellulose, ash content, hot water and 1% NaOH solubilities) were determined according to relevant TAPPI test, Kurscher–Hoffner and Chlorite methods. Meanwhile, fibre dimension and pulp properties were measured after the pulping process. The mechanical strength of handsheet produced (tensile, burst, tear and fold) was investigated according to the TAPPI test method. Scanning Electron Microscope (SEM) was used to observe and determine the morphological characteristic of the handsheet surface. In order to propose the suitability of the studied plants as alternative fibre resources as pulp in papermaking, the obtained results are compared to other published literatures from wood resources. Results show that the lignin (5.67%), hot water (3.83%) and 1% NaOH (19.64%) solubility contents of cogon grass are the lowest compared to cassava peel, cocoa pod husk and oil palm leaf. The contents have influenced the production of the highest pulp yield which is 35.68%. Although cogon grass contains shorter fibre than oil palm leaf, the handsheet product showed the highest tensile (45.06 Nm/g), burst (3.90 kPa.m^2/g) and tear (2.17 mN.m^2/g) indices when compared to oil palm leaf (12.08 Nm/g, 0.95 kPa. m^2/g and 1.80 mN.m^2/g) and published wood resources. From SEM images, handsheet of cogon grass contains compact, straight and smooth fibres. In conclusion, apart from the chemical, pulp, physical and mechanical properties and the surface morphology of the cocoa pod husk, cogon grass and oil palm leaf sheets indicate that they are suitable to be used as alternative fibres for pulp and paper-based industries with cogon grass being the best.

ABSTRAK

Permintaan global terhadap serat kayu semakin meningkat sejajar dengan peningkatan penduduk dan penghasilan baru daripada serat kayu. Oleh itu, untuk membekalkan serat bukan kayu telah diperkenalkan sebagai alternatif lain dalam industri pembuatan pulpa dan kertas. Kajian ini bertujuan untuk menggunakan serat bukan kayu sebagai pulpa dalam industri pembuatan kertas: mempromosikan konsep "waste to wealth" dan "bahan kitar semula". Oleh itu, objektif kajian ini untuk mengenal pasti potensi kulit ubi kayu, sekam pod koko, lalang dan daun kelapa sawit sebagai serat alternatif dalam pembuatan pulpa dan kertas berdasarkan kepada ciri-ciri kimia, pulpa, fizikal dan kekuatan mekanikal. Sifat-sifat kimia yang terlibat dalam kajian ini (holosellulose, selulose, hemisellulose, lignin, abu, kelarutan air panas dan 1% NaOH) melalui kaedah yang releven iaitu TAPPI, Kurscher-Hoffner dan kaedah klorida. Manakala, dimensi serat dan ciri-ciri pulpa ditentukan selepas proses pemulpaan. Kekuatan mekanikal kertas (tegangan, pecahan, koyakan dan lipatan) telah diuji melalui kaedah TAPPI. Mikroskop elektron imbasan (SEM) telah digunakan untuk mengkaji morfologi di atas permukaan kertas. Dalam mengetengahkan kesesuaian kajian ini sebagai alternatif dalam penghasilan serat pulpa dalam pembuatan kertas. Keputusan kajian ini dibandingkan dengan kajian terdahulu terutamanya dari sumber kayu bagi memastikan kesesuaian serat bukan kayu di dalam pembuatan pulpa dan kertas. Keputusan menunjukkan bahawa lalang mengandungi lignin (5.67%), keterlarutan air panas (3.83%) dan 1% NaOH (19.64%) yang rendah dibandingkan dengan kulit ubi kayu, sekam pod koko dan daun kelapa sawit. Jumlah ini mempengaruhi kepada penghasilan pulpa yang tinggi (35.68%). Walaupun lalang mengandungi serat yang pendek daripada daun kelapa sawit, tetapi produk kertas yang telah dihasilkan mempunyai nilai yang tinggi dari segi tegangan (45.06 Nm/g), pecahan (3.90 $kPa.m^2/g$) dan koyakan (2.17 $mN.m^2/g$) dibandingkan dengan daun kelapa sawit (12.08 Nm/g, 0.95 $kPa.m^2/g$ dan 1.80 $mN.m^2/g$) dan sumber dari kayu. Daripada imej SEM menunjukkan bahawa lalang

mempunyai serat yang padat, lurus dan lembut. Kesimpulannya, sebahagian daripada ciri-ciri kimia, pulpa, fizikal dan kekuatan mekanikal serta sifat morfologi permukaan sekam pod koko, lalang dan daun kelapa sawit menunjukkan kesesuaian sebagai sumber serat gentian dalam industri berasaskan pulpa dan kertas, yang mana lalang menunjukkan potensi yang paling baik.

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LIST OF SYMBOLS AND ABBREVIATIONS

D	-	Fibre diameter
L	-	Fibre length
%	-	Percentage
^{o}C	-	Degree Celsius
$<$	-	Less than
$>$	-	More than
1% NaOH	-	One percent of sodium hydroxide
CO_2	-	Carbon dioxide
$C_2H_4O_2$	-	Acetic acid
C_2H_6O	-	Ethanol
$(C_6H_{10}O_5)n$	-	Cellulose formula
CH_3COCH_3	-	Acetone
ClO_2	-	Chlorine dioxide
H_2O	-	Water
H_2O_2	-	Hydrogen peroxide
H_2S	-	Hydrogen sulfide
H_2SO_4	-	Sulfuric acid
HNO_3	-	Nitric acid
Na_2S	-	Sodium sulfide
Na_2CO_3	-	Sodium carbonate
$NaClO_2$	-	Sodium chlorite
NH_3	-	Ammonia
SO_2	-	Sulfur dioxide
AFM	-	Atomic Force Microscopy
AFPA	-	America Forest and Paper Association
AQ	-	Anthraquinone
B_I	-	Burst index

BE	-	Back-scattered electron
CD	-	Cross machine direction
Cell	-	Cellulose
Eq.	-	Equation
F	-	Force
F_E	-	Folding endurance
F_D	-	Fibre diameter
F_L	-	Fibre length
FRIM	-	Forest Resources Institute Malaysiaha
ha	-	Hectares
Hemi	-	Hemicellulose
Holl	-	Holocellulose
Hw	-	Hot water solubility
ISO	-	International Organization for Standardization
IT	-	Information Technology
JPM	-	Jabatan Perangkaan Malaysia
Lig	-	Lignin
MC	-	Moisture content
MD	-	Machine direction
MOA	-	Ministry of Agriculture and Agro-based Malaysia
MPOB	-	Malaysia Palm Oil Board
n.a	-	Non-available
NaOH	-	Sodium hydroxide
$NaClO_2$	-	Sodium chlorite
o.d	-	Oven-dried
PITA	-	Paper Industry Technical Association
RH	-	Relative humidity
rpm	-	Rotation per minutes
SE	-	Secondary electron
SEM	-	Scanning Electron Microscope
SPSS	-	Statistical Package for Social Science
T_I	-	Tensile index
T_{EI}	-	Tear index
TAPPI	-	Technical Association of Pulp and Paper Industry

TRS	-	Total reduced sulfur
USA	-	United State of America
UTHM	-	Universiti Tun Hussein Onn Malaysia
WBG	-	World Bank Group

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CHAPTER 1

INTRODUCTION

1.1 Background of study

Historically, non-woody plants were major resources for pulp and paper production compared to woody plant. In the 19th century, an insufficient supply of the traditional raw materials of cotton and linen rags made it necessary to use wood to make paper (Bajpai *et al.,* 2004). Today, wood fibre either hardwood or softwood are the main raw materials used for the production of pulp and paper. About 90-92% of the pulp and paper in the world are produced from wood, mostly in the developed countries (Bajpai *et al.,* 2004 and Jiménez *et al.,* 2009) such as Canada and United State of America, USA (Madakadze *et al.,* 2010).

Generally, the pulp and paper industries obtain cellulose from hardwood or softwood (Sridach, 2010a). Jean and Santosh, (2006) reported that the total capacity of pulp and paper production from wood resources is more than one million tons/year. However, this amount is higher in Malaysia due to the increasing pulp production and consumption in paper-based products (Figure 1.1). In 2007, the net paper consumption in Malaysia is approximately three million metric tons (Goyal, 2010a).

The demand of paper consumption was increased in 2009 when the average of paper consumption was about 151 kg/capita (Katrin, 2010) and this value is expected to reach to 200 kg/capita by the year 2015. In fact, Malaysia presents the third highest amount of paper consumption in the world together with Korea, Taiwan, Singapore and Hong Kong as presented in Figure 1.2. Moreover, Malaysia also ranked the second highest demand of paper consumption in Asia region (Katrin, 2010). Malaysia is a developing country, where the high demand of paper consumption is due to the increase in population, lack of awareness of environment chain effect and less practice in paperless document (intact with technology) (Jean and Santosh, 2006).

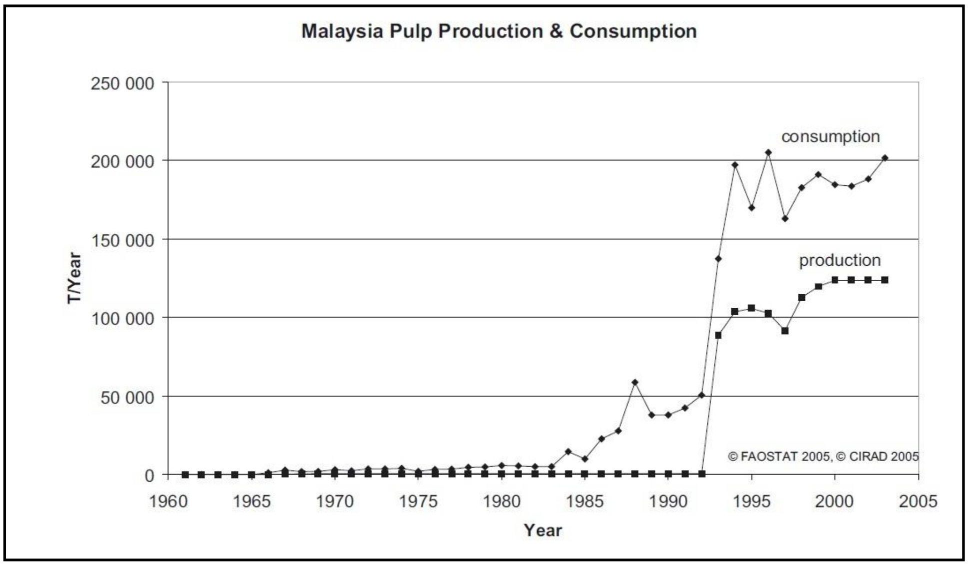

Figure 1.1: Malaysia's pulp production and consumption (Jean and Santosh, 2006)

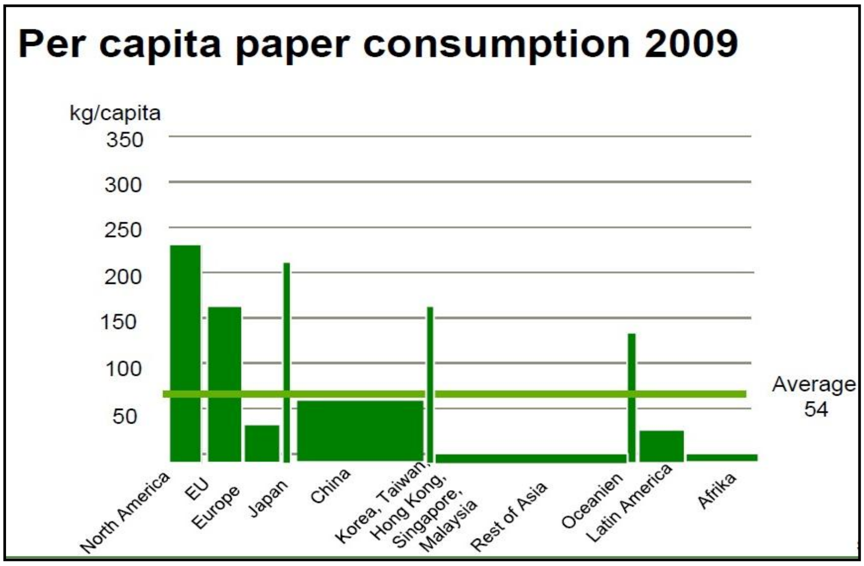

Figure 1.2: Paper consumption in Malaysia and worldwide (Katrin, 2010)

Owing to the environmental concerns and wood resource depletion, more attention is being paid to renewable materials such as non-wood. Conventionally, wood is the principal resource of cellulosic fibre for pulp and paper manufacture. The increase demand of paper consumption from virgin pulp is the main cause for the usage of wood species as the main raw material leading to massive deforestation and replantation. This has consequently altered the ecological balance and contributed to the climate change. The use of non-wood fibre resources through rational and innovative ways of utilisation can be considered as new alternative cellulosic fibre resources. The use of non-wood could promote the concept of “from waste to wealth” and “recyclable material” which is aimed to build a sustainable and sound material-cycle society through the effective use of resources and green technology.

The non-wood fibres could be obtained from three sources: agricultural by-product, industrial crops and naturally growing plants (Sridach, 2010b). Agriculture is an important sector in Malaysian economy. Agricultural wastes/by-products have been shipped away for processing or disposing after post-harvest. Diversification of the industries is crucial in encouraging economic stability while growth value-added processing would help in agricultural utilisation.

The concept of recycling has been around for a more appealing reason these days. Recycling has always been implemented as a way to reuse materials that are no longer needed or suitable for initial purpose or use (Hecker, 2005). For instance, cocoa and cassava plants are important products in Malaysia, used in producing cocoa powder and cassava crisp respectively. Thus, at every cocoa and cassava harvesting season, large quantities of cocoa pod husks and cassava peels become abundantly available but discard as wastes. Another plant that presence in abundance in Malaysia is cogon grass. It has short cycle growth compared to wood plants. In addition, it is also an invasive species that causes problems for livestock and wildlife. Oil palm leaf are also an agricultural residues obtained from oil palm plantation. These wastes are abundant and create problems to farmers for disposal.

Normally, in paper-based industry, pulping is compulsory. Pulping is a process of extracting cellulosic fibres from plant material, generally hardwood, softwood trees or non-woody plants. The most abundant component of the native wood matrix is cellulose, a polysaccharide that is desired for paper production (Rowell *et al.,* 2000). Commonly, pulping process could be conducted through three types which are chemical, mechanical (including thermomechanical) and semi-chemical pulping processes (Sridach, 2010b).

Mechanical pulping process separates fibres from each other by applying mechanical energy applied to the wood matrix, causing the gradual break of the bonds between the fibres and also release of fibre bundles, single bundles and fibre fragments. Mechanical pulps are weaker than chemical pulps, but cheaper to produce

(50% of the costs of chemical pulp and generally obtained in the yield range of 80–95%) (Bajpai, 2012). Semi-chemical process involves mechanical abrasion and the use of chemicals (Biermann, 1996a). This process is ideally suited for the production of pulp yield between 60-80% (Veguru and Cameron, 2005).

Chemical pulping is used on most papers produced commercially in the world today. Traditionally, this has involved a full chemical treatment to remove non-cellulose materials or other lignocellulosic plants components leaving intact the cellulose fibres (Bajpai, 2012). Yields of chemical process are on the order of 40 – 50% and produce high strength in paper-based product compared to mechanical and semi-chemical processes (Charbonneau *et al.,* 1994 and WBG, 1998).

In this research, the pulping process chosen is chemical process as it contributes to higher paper strength produced. In chemical process, two types are available: alkaline and acidic processes. This pulping also dissolves the lignin (act as glue that holds plants fibre) from cellulose and hemicelluloses in raw material (Biermann, 1996a). Although it produces less amount of yields compared to the mechanical pulping but the quality of yield is better than other pulping processes where the paper shows higher strength in tearing and tensile (Sridach, 2010b).

Therefore, the alkaline and acidic pulping processes at constant variables (temperature, cooking time and concentration of acid or alkaline) are investigated in this study to produce handsheets from non-wood materials. Subsequently, the properties of handsheet produced from each sample (cassava peel, cocoa pod husk, cogon grass and oil palm leaf) will be evaluated through the paper testing and strength properties (tensile, tearing and bursting strength and folding endurance).

It is hypothesized that the alternative non-wood has fibre content that are comparable to that of conventional wood. This study provides valuable information of introducing non-wood raw materials to be utilised in the paper-based industry in Malaysia, hence reassuring the sustainability use of natural resources.

1.2 Problem statement

Paper is becoming an important commodity of today's society. The pulp and paper industries have been rising due to the increased demand of paper-based products. The consumption of paper has also been steadily increased over the world. With the advent of information technology (IT), the world paper consumption was expected to decrease with increasing deployment of paperless communication. However, the opposite has instead happened. The paper consumption in Malaysia has tremendously increased since 1960 to 2005 as shown in Figure 1.3 (Jean and Santosh, 2006).

The paper consumption is higher than the paper production in the years of 1960 to 2005. Besides, Malaysia's consumption of paper products increase continuously due to several reasons, such as population growth, better literacy and industrialisation in developing this country (Enayati *et al.*, 2009). For example, the increase of paper utilisation in Figure 1.3 is parallel to the population growth which is 27,058,000 to 28,964,000 people in 2007 to 2011 (JPM, 2012). Moreover, the scarcity of wood resources as major cellulosic for pulp and paper industry has caused the lower amount of paper product as shown in Figure 1.3.

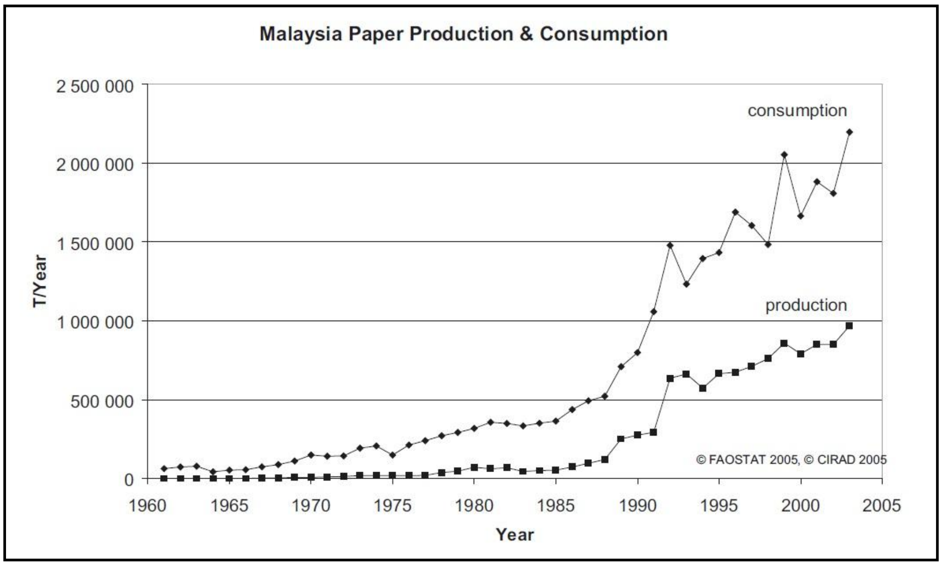

Figure 1.3: Malaysia's paper production and consumption (Jean and Santosh, 2006)

In Malaysia, the depletion of wood as main resource in pulp and paper-based industry is due to the massive scale of deforestation. Deforestation is a sensitive issue in Malaysia as its rate is accelerating faster than any other tropical countries in the world as can be seen in Table 1.1 (Rhett, 2005). Table 1.1 shows that Malaysia's annual deforestation rate jumped almost 86% for the years of 1990 to 2005. In addition, Malaysia showed the total average of lost of forest around 140,200 ha or 0.65% of its forest area every year since 2000. In Malaysia, around 124,000 metric tons of pulp for paper were obtained from wood resources that provide 184,000 metric tons of paper in 2002 (Rhett, 2005). However, this amount still does not enough to fulfil the domestic consumption.

Table 1.1: Rate change in total deforestation rate between 2000-2005 period versus 1990-2000 period (Rhett, 2005)

Country	**Rate of change (%)**
Malaysia	85.7
Cambodia	74.3
Burundi	47.6
Togo	41.6
Nigeria	31.1
Sri Lanka	25.4
Benin	24.1
Brazil	21.1
Uganda	21.0
Indonesia	18.6

Deforestation is known as clearing Earth's forests on a massive, frequently resulting harm to the quality of the land. The world's rain forest especially in Malaysia could completely disappear in a hundred year at the current rate of deforestation (Rhett, 2005). Normally, forests are cut down for many reasons, but most of them are related to fulfil the demands of people such as pulp and paper products. As the consumption of paper in Malaysia increase quickly, there is a huge enhance in the demand of wood fibre resources for paper production. Therefore, many forest areas are cleared in order to fulfil the demand of paper products. Nowadays, logging operation regarding Malaysia's wood and paper products, has cut countless trees every year. In addition, illegal loggings were also reported where roads are built as access to more remote forests which in turn lead to further deforestation (Elias, 2011).

Generally, deforestation has created many negative effects on the environment and it causes various problems in Malaysia and the world. The main dramatic impact is the loss of habitat for millions of species. There are many animals and plant species that live in the forests and many from these species cannot survive after deforestation as it destroys their habitats. Besides, deforestation also causes climate change. Normally, forest soils are moist, but they are quickly dry out due to the direct sun ray without protection from the tree cover. In addition, trees are also important because they help to carry on the water cycle by returning water vapour back into the atmosphere. Without trees to fulfil these functions, many former forest lands could become deserts in the near future.

Today, recycled paper plays important materials in pulp and paper industry in Malaysia. In the light of the shortage of wood plants, the cost effective and abundance of recycled paper make the recycled paper a reasonable candidate for pulp and papermaking in this country. Unfortunately, 100% of recycled paper cannot be used in paper production because of the strength of recycled fibre has decreased during drying and rewetting cycle. This situation happens due to the phenomena of irreversible hardening or hornification of fibres which causes the fibre to lose their conformability and swelling capacity (WanRosli *et al.,* 2005).

In addition, hornification occurs when the hydrogen bonds that are formed between cellulose chains in the wall during resistance are being broken during rewetting process (WanRosli *et al.*, 2005). As to solve this problem, the world is seeking for the alternative resources to substitute the virgin fibre in paper-making industry.

Non-woods are commercially used in countries that have limited of origin resource industry. China and India use non-woody plants (bamboo, jute, rice straw and bagasse) as alternative fibres in their paper production (Jahan *et al.*, 2007). Therefore, the alternative fibre resources from agricultural wastes are great to be used in pulp and paper-based industries in Malaysia as it also eliminates the problem of solid waste disposal. Agricultural waste is also known as organic waste. Therefore, this study suggests four types of organic wastes: cassava peel, cogon grass cocoa pod husk and oil palm leaf as alternative fibres in paper making industry.

Agriculture is an important economy sector in Malaysia. Cassava and cocoa are the economical plants in this country. These crops have been produced around 68,508 metric tons (MOA, 2011) and 7,019 metric tons (JPM, 2011) in 2010 respectively. Therefore, these crops will generate high amount of solid wastes after they have been processed. These solid wastes (agricultural wastes) have been shipped away for processing or disposing at landfills. The abundance of solid wastes produced from these plantations has created great environmental problems in Malaysia (Reddy and Yang, 2005).

On the other hand, cogon grass is a rapidly growing perennial grass that is widely found in Malaysia. This invasive grass could causes problems to plantation areas. In addition, cogon grass is very much flammable and can burn even when it is still green. Dense stands of dry cogon grass burn intensely in hot temperature and it kills other vegetation around them (Jennings *et al.*, 2012). Oil palm trees have also become one of the most valuable commercial crops in Malaysia, with around 4.85 million ha of plantation area in 2010. With a large plantation area, they create abundance of oil palm leaves as one of the oil palm biomass residues (Sulaiman *et al.*, 2012). Therefore, these leaves can also be suggested as alternative fibre in pulp and paper-based industries.

Chemical pulping especially kraft and sulfite processes provide a lot of bad effects to the environment because of the high generate emission of sulphur dioxide, SO_2 and wastewater pollution (Li *et al.*, 2012 and Sridach, 2010b). In industrial scale, big amount of chemical have to be used to produce pulp and paper-based and it also generate a lot of chemical waste after processing. This chemical waste must be treated before it is release back to environment.

Therefore, cassava peel, cogon grass, cocoa pod husk and oil palm leaf are studied as alternative fibres in pulp and paper-based industry. Furthermore, the use of these materials could reduce the massive scale of deforestation and environmental problems in Malaysia.

1.3 Research aim

The aim of this project is to investigate the potential of non-wood plant resources: cassava peel, cocoa pod husk, cogon grass and oil palm leaf as fibre substitution to the wood resources and conventional imported virgin pulp in the paper-making. The experimental investigation is complemented by different types of chemical pulping process (alkaline and acidic processes) at constant variables of pulping conditions.

1.4 Research objectives

The specific objectives of this project are:

i. To investigate the chemical and physical properties of cassava peel, cocoa pod husk, cogon grass and oil palm leaf.

ii. To determine the effects of two types of chemical pulping (alkaline and acidic processes) with the constant variables (cooking time, active alkaline or acidic, temperature and liquor to solid ratio) of cassava peel, cocoa pod husk, cogon grass and oil palm leaf to produce the good pulp yield and quality pulp for handsheet making.
iii. To determine the mechanical properties of handsheets from cocoa pod husk, cogon grass and oil palm leaf which are focused on tensile and bursting strength, tearing resistance and folding endurance as important parameters in selecting alternative fibre resources.
iv. To observe the surface morphological of handsheets made from cocoa pod husk, cogon grass and oil palm leaf.

1.5 Scope of study

This study focuses on four different types of non-wood fibre resources: cassava peel, cogon grass, cocoa pod husk and oil palm leaves as alternative fibres to wood fibre resources in pulp and paper-based industries. These samples were collected around Batu Pahat, Johor. The samples used were in the form of particles (0.40 - 0.45 mm) and air-dried (2 - 5 cm long) for the study of their chemical properties, physical properties, morphology characterisation and pulping process respectively (Figure 1.4).

The properties of non-woods, both chemical and physical properties are important variables to indicate the suitability of samples to substitute wood fibre resources in pulp and paper-based industries. For determination of chemical properties, the scope was on cellulose, hemicellulose, holocellulose, lignin, 1% NaOH solubility, hot water solubility and ash content. Meanwhile, in term of physical properties the focus was limited to fibre lengths and diameters of the samples only.

Furthermore, since chemical process was chosen for the pulping process in this study, the effects of different types of chemicals (alkaline and acidic) were also analysed at constant variables based on the screened pulp yield of these samples.

At the end, the handsheet of samples were formed after each alkaline and acidic process. It is an important procedure to measure the strength of samples in order to produce good quality handsheets and a good indicator whether or not it can be utilised as alternative fibres in the paper-based industry. Hence, the strength of the handsheet was investigated in term of tensile and bursting strength, folding endurance as well as tearing resistance. In addition, surface morphology structure was also studied using handsheet of samples which focused on the cross-section and surface of handsheet.

Results obtained from each experiment were triangulated and analysed before the conclusion on the best alternative fibre was made. The relationship between all scopes in this study is shown in Figure 1.4.

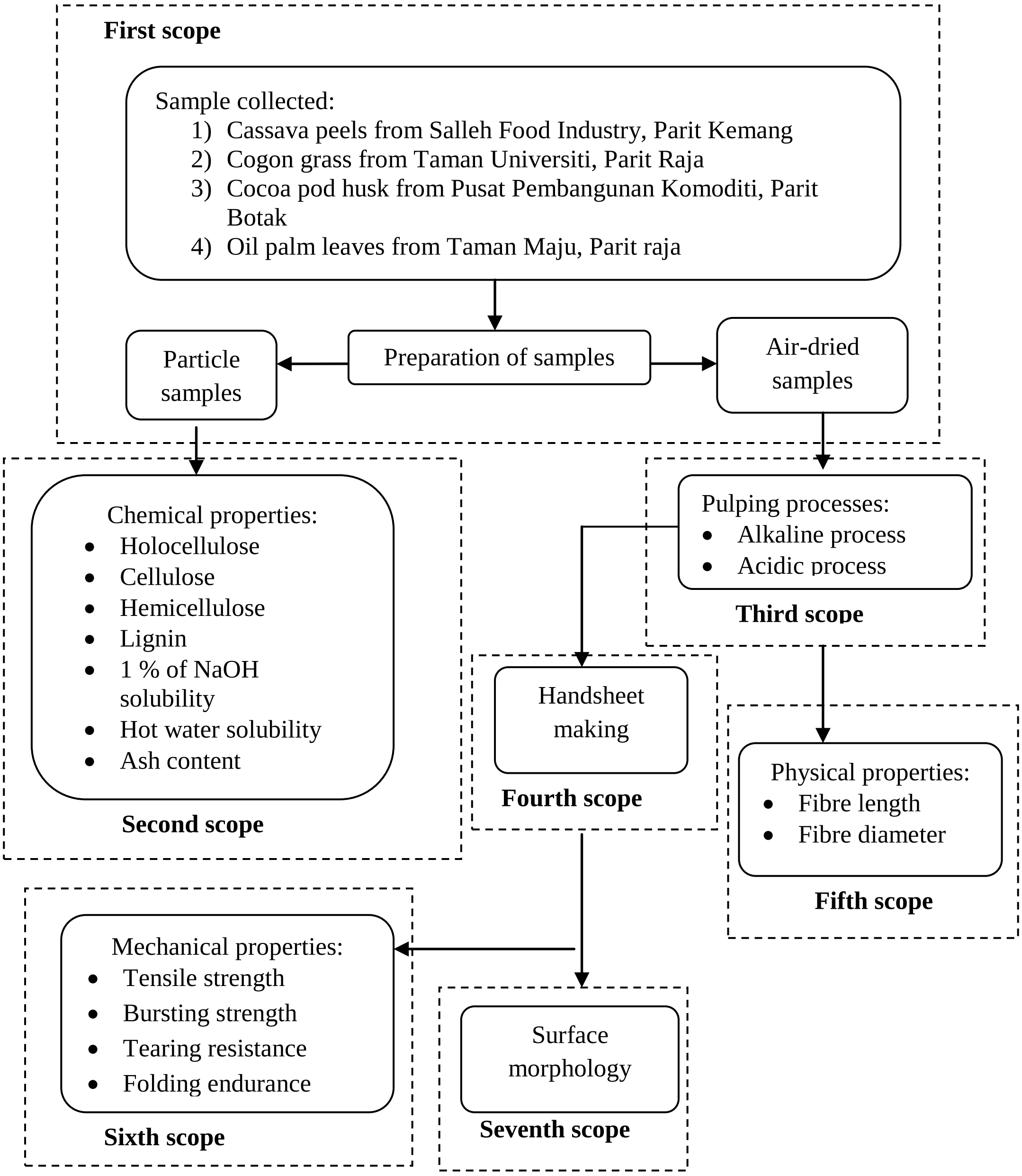

Figure 1.4: Flow diagram of the relationship between the project scopes

1.6 Significance of study

To bridge over the extensive gap between demand and supply, many non-wood biomass have been recognised and investigated to appraise their suitability for pulp production. Therefore, this study focuses on non-woody plant materials including agricultural wastes: cassava peel, cogon grass, cocoa pod husk and oil palm leaf as alternatives to the increasingly scant forest wood as the major resource for pulp fibre in paper-based industry. The principal interest in pulping non-woody raw materials is that they provide pulp of excellent quality for making specialty graded paper or constitute the sole affordable source of fibrous raw material in some geographical areas.

Normally, production in pulp and paper-based industries involves massive felling of trees, which in turn leads to deforestation. Rapid increase of competition for wood supplies coupled with slowly rising cost of wood have generated renewed interest in the use of non-wood plant fibres for paper-making in the highly industrialised countries. The use of agricultural wastes in pulping and paper-based industries might be advantageous because it prevents the need for disposal, which currently increases farming costs and causes environmental deterioration through pollution and fires.

In addition, increasing the reuse of the non-wood fibres may bring significant rational in reduction of wood consumption and could allow preserving the forestry resources as well as a positive impact on the environment problems. Moreover, new substances from non-wood resources may allow modification of pulp and paper properties to produce better quality paper products.

The alternative fibres also provide good quality of paper production comparable with the wood resources in paper-based. In addition, alternative fibres also increase the performance in recycled paper by increasing the strength properties with combination of fibres during pulping process. The alternative fibres also encourage the application of green technology in terms of generating new paper production from non-wood resources to create more environmentally friendly processes. This research is conducted to suggest alternative fibres from agricultural wastes as pulp in paper-based industries. In the long run, environmental issues such as deforestation and chemical waste pollution could be reduced in Malaysia.

1.7 Summary of thesis chapters

Chapter 1 provides an overview of this study to find suitable alternative fibre in pulp and paper-based industries. This chapter describes about background, problem statements, objectives, scope and significance of study.

Chapter 2 presents theory and a literature review for the study into non-woody plants, setting this project in the context of a wider body of knowledge. This chapter covers include non-woody materials, chemical, physical and mechanical properties, chemical pulping process, and experimental theory used in this study.

Chapter 3 presents the experimental equipment and methodology for the project. The chapter describes how the non-woody plants were prepared, the chemical and physical properties of the pulp sample testing. The method used for mechanical testing and verification is explained. Finally, the approach for the surface structure handsheets is described.

Chapter 4 provides detailed results and discussions from the experimental investigation. Data was collected at different testings and compared to previous studies. The overall findings of the studied materials are presented.

Chapter 5 summarises the main conclusion resulting from this study and recommendations for further works are discussed.

Appendices are attached with supplementary materials. Data on the materials of chemical, physical and mechanical properties distributions are provided in Appendix A. The statistical analyses for all parameters in this study are attached in Appendix B. The flow diagram on the pulping process and handsheet making of all materials are provided in Appendix C. Appendix D is the paper publications relating to this study.

CHAPTER 2

LITERATURE REVIEW

2.1 Introduction

This chapter describes the theory and literature that are relevant to this study on non-wood resources as alternative fibres in pulp and paper-based industries. This chapter proceeds by describing the overview of pulp and paper manufacturing and fibre resources involved in the industry worldwide (Section 2.2 and Section 2.3 respectively). The goal of finding alternative fibres in pulp and paper-based industries from non-wood plants is elaborated in Section 2.4.

The fundamental properties of non-wood plants materials: their chemical and physical properties are described in Section 2.5 and 2.6 respectively. The use of chemical liquor for removing lignin polymer and producing single fibre is described in Section 2.7. Section 2.8 and 2.9, these are the most important sections in this study where their most desirable goal is to determine the suitability of non-wood plants materials used as an alternative fibre in pulp and paper-based industries.

2.2 Overview of pulp and paper manufacturing

Paper is essentially a sheet of cellulose fibres with a number of added constituents to affect the quality of the sheet and its fitness for intended end use. The term of paper generally refers to the weight of the product sheet (grammage) with paper ranging up to 150 g/m^2. In paper production, the fibres are usually vegetative but mineral, though animal or synthetic fibres can also be used (Alexandersson, 2003). The name paper originates from the Greek and Roman word for papyrus, which was a sheet made from thin sections of reed (*Cyperus papyrus*). Paper was also used in ancient

Egypt from this paprus (Holik, 2006). Today's kind of paper was first developed and used in China since 1990 (Zhuang *et al.,* 2010).

Nowadays, paper has transformed from an uncommon artisan material to a commodity product, with a high practical value in communication, educational, and technical applications (Holik, 2006). The general term of paper refers to all products that are produced in the paper industry. They can be further divided into four main categories: paper, tissue, paperboard and speciality papers (Dick *et al.*, 2006). Paper category refers to the paper for writing, printing and copying that are classified as either wood-free or wood containing. Paper products was made from at least 90% chemical pulp are wood-free paper whereas wood-containing paper is refers to bleached mechanical pulp (Alexandersson, 2003).

The second category is tissue product such as paper towels, handkerchiefs and napkins. The third category is paperboards, where they are usually used for different packaging products and can be further divided into cartonboards, containerboards and special boards. As comparison between papers with paperboards, paper is usually thinner, lighter and more flexible than paperboards. The last category is specialty paper where they are different paper products like filter paper, electrical insulation paper for cables, coffee filters and tea bag papers (Alexandersson, 2003).

The worldwide consumption of paper is increasing steadily over the years. The global pulp and paper industries is dominated by North American (United States, Canada), northern European (Finland, Sweden) and East Asian countries such as Japan (Bajpai, 2012). The paper consumption worldwide amounted to roughly 371 million tonnes in 2009 (Cielo, 2011), where 40% of this paper consumption belonged to Asian region (Bajpai, 2012). In Asia region, Japan is the lead consumer of paper products per capita followed by Malaysia and Singapore (Katrin, 2010). Figure 2.1 shows the paper consumed in the global and they are divided into five major segments of end-use which are printing and writing paper, newsprint, tissue, container board and other paper and paperboard (Dick *et al.,* 2006).

From Figure 2.1, the capacity production of paper segments is very high because the paper companies normally run continuously. This is due to fulfil the demand of paper segments consumption (Dick *et al.,* 2006). The ratio of the worldwide consumption of different paper and paperboards has changed in the past and will change in the future according to technical and social evolution and developments in the individual countries and world as a whole (Cielo, 2011).

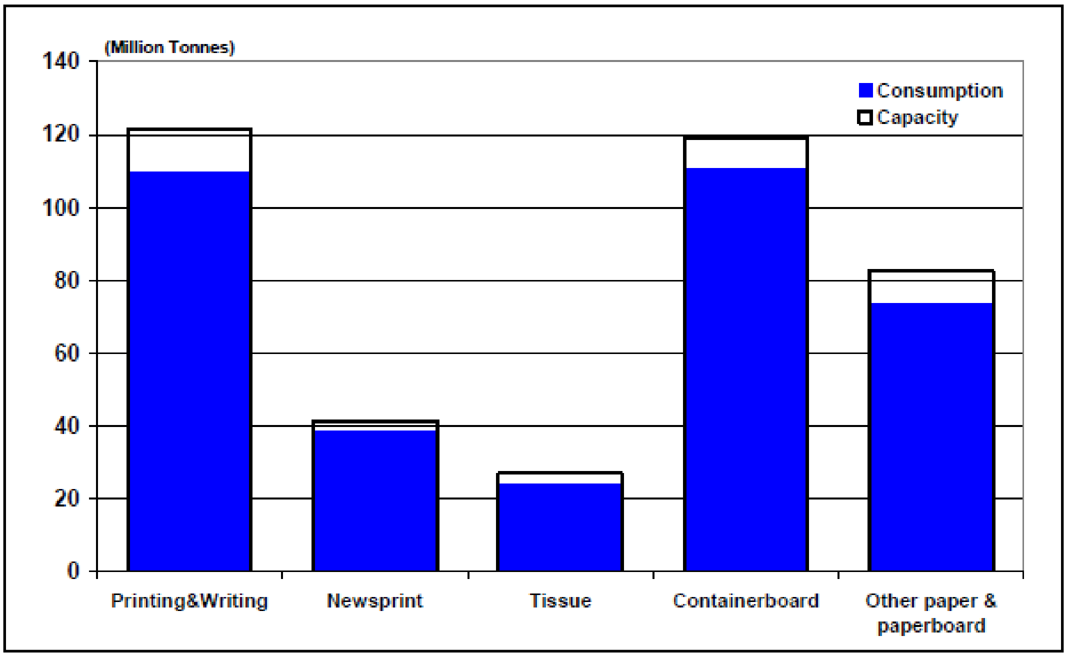

Figure 2.1: Consumption and capacity for the year 2004 within different paper segments (Dick *et al.*, 2006)

The components used in paper and paperboard production worldwide are given in Figure 2.2. Today, recovered paper has become the lead resource for paper and paperboard production, followed by chemical pulp, mechanical pulp, pigments and fillers and chemical additives (AFPA, 2010 and Holik, 2006). Paper is mainly based on fibres from wood, renewable and recyclable raw materials. The specific characteristics of these fibre materials are that the paper strength results from the hydrogen bonding between the individual fibres. The pulps produced in different ways have different properties, which make them suitable for particular products. Most pulp is produced for the purpose of subsequent manufacture of paper or paperboard. Some is destined for others such as thick fibreboard or products manufactured from dissolved cellulose (Holik, 2006).

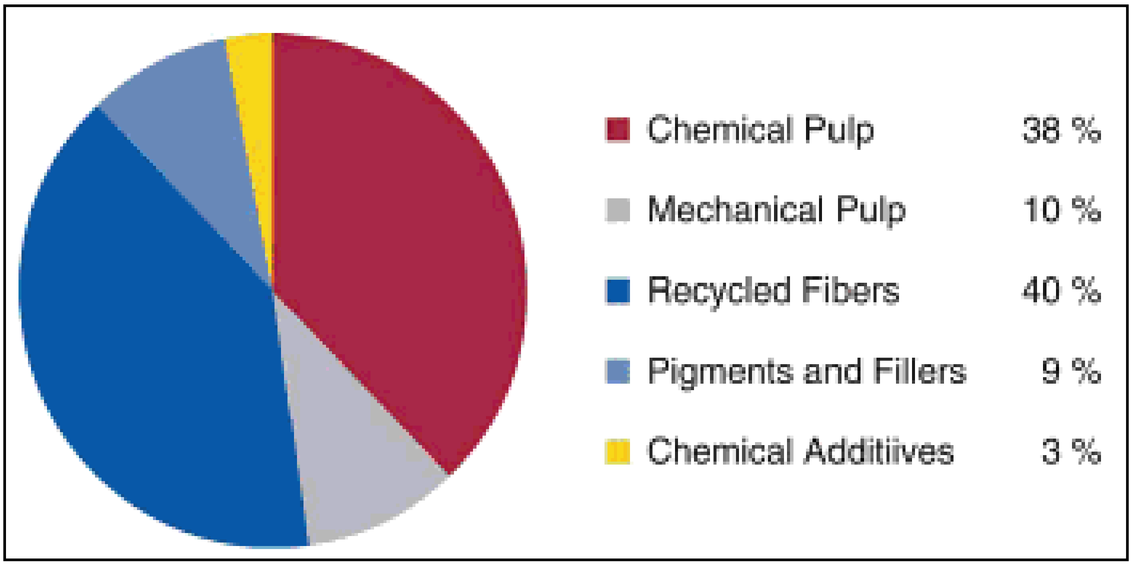

Figure 2.2: Components used in paper and paperboard production worldwide (by mass ratio) (Holik, 2006)

2.3 Fibre resources for pulp and paper

Normally, fibre resources for pulp and paper are obtained from trees or agricultural crops. These resources include plant materials harvested directly from the land (wood, straw and bamboo), plant material by-products or residual from other manufacturing processes (wood chips from sawmills, bagasse and cotton linear) and fibre recovered from recycled paper or paperboard. Forest resources have important value in producing a range of different wood resources for pulp and paper-based industries (Holik, 2006).

Wood resources are divided into two types which are softwood (such as spruce, pine, fir, larch and hemlock) and hardwood (such as eucalyptus and birch). Huge majority of wood resources (more than 90-92% of fibres) are used for pulp and paper production globally (Jiménez *et al.*, 2009 and Sridach, 2010a). These wood resources are used in many kinds of paper grades due to its smooth surface area and strong strength (Dick *et al.*, 2006).

Table 2.1 shows the comparison of chemical composition between two types of wood resources. Wood consists mainly of cellulose, hemicellulose, lignin, extractives and ash. The chemical composition of wood resources varies from species to species (Henricson, 2004). In general, hardwoods have higher cellulose content (43 - 47%) and lower lignin (16 - 24%) and extractives (3 - 8%) contents as compared to softwoods (cellulose 40 - 44%, lignin 25 - 31% and extractives 10 - 25%).

Table 2.1: Chemical properties in hardwood and softwood resources (adopted from Henricson, 2004 and Koch, 2006)

Wood types	Cellulose (w/w %)	Hemicellulose (w/w %)	Lignin (w/w %)	Extractives (w/w %)	Ash (w/w %)
Softwood	40 - 44	25 - 29	25 - 31	10 - 25	0.2 - 0.4
Hardwood	43 - 47	23 - 35	16 - 24	3 - 8	0.2 - 0.8

Trees needed to meet virgin wood fibre demand of the forest product industry are already growing except for the new fast growing plantations. Therefore, in global term, there will not be a long-term fibre shortage. However, fibre supplies within and across particular regions will tighten. These regional imbalances are already significant and will continue to grow. From Figure 2.3, Asia is presently the largest fibre deficit region, followed by Western Europe. At the same time, Asia is the focus of fibre demand growth for pulp and paper. If this assessment is accurate, pulp and paper industry’s dependence on virgin fibres must be reduced by expansion in the use of recovered paper and growth in the use of non-wood plant fibre in Asia (Chandra, 1998).

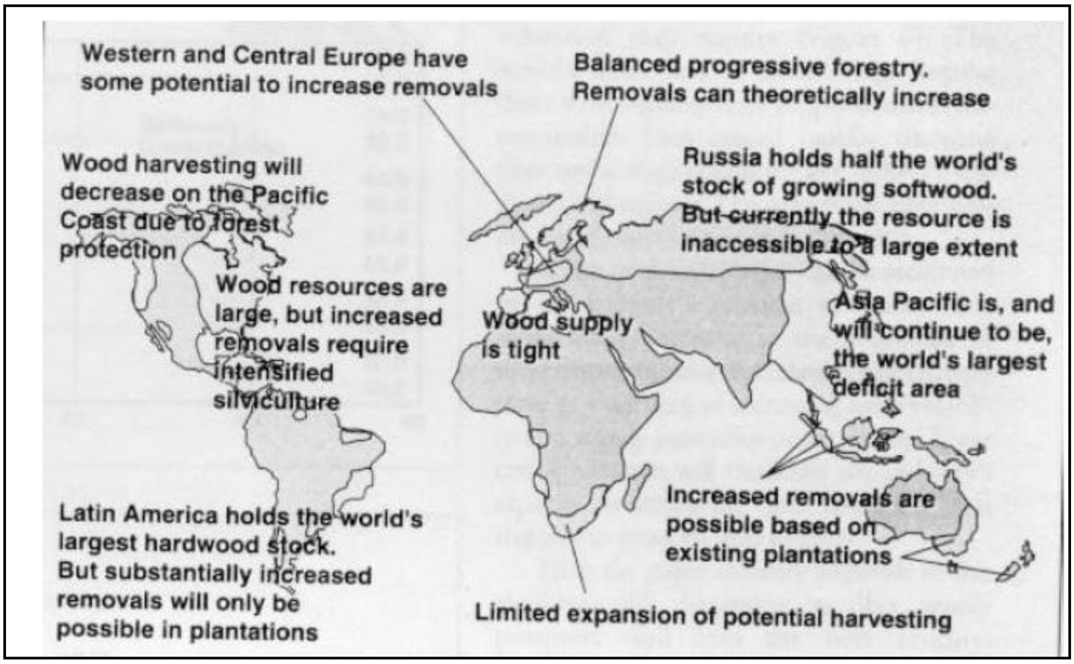

Figure 2.3: Main global forest resources and wood deficit areas (Chandra, 1998)

Nowadays, many other countries are looking for non-wood plants fibre resources as alternative fibres in pulp and paper-based industries. This is due to the depletion and rising prices of wood resources and readily available non-wood fibre resources in these countries (Atchison, 1992). The United States is also looking for non-wood fibres to be resources of pulp and paper-based as alternative fibre in this industry to replace the virgin fibre resources (Ai and Tschirner, 2010). At the same time, Europe has a shortage of short fibre hardwood pulp and is thus an importer of this kind of pulp (Lopez *et al.*, 2011). They found out that some of the non-wood fibres have the properties to replace these fibres. Hence, the use of non-wood fibre for pulp and paper-making is thus also expected to grow in Europe (Chandra, 1998). Indeed, China and India are the lead countries that use of non-wood fibres in pulp and paper production rather than other countries in the world (Mabee and Pande, 1997).

The total of non-wood plants (8 - 10%) pulping capacity worldwide is increasing faster than the wood pulping capacity (González *et al.*, 2008 and Rodríquez *et al.*, 2008). In China, the consumption of non-wood resources in pulp and paper-based industries is higher than wood sources from the year 1995 to 2005, as can be seen in Table 2.2 (Sbrilli, 2007). The development of these industries will need a continuous and sustainable forestry around the world. This is also due to the fact that non-wood plants sources have displayed different kinds of advantages in pulp and paper-based production compared to the wood resources as presented in Table 2.3.

Table 2.2: Consumption amounts for the most common raw materials in the Chinese paper industry from years 1995 to 2005 (Sbrilli, 2007)

Resources	**Pulp consumption (thousand tons)**		
	1995	**2000**	**2005**
Non-wood	11,360	11,150	12,600
Wood	2,830	5,350	11,300

Table 2.3: Comparison of non-wood and wood resources for pulp and paper-making (adopted from Rousu *et al.*, 2002 and Kissinger *et al.*, 2007)

Description	Fibre resources	
	Wood	**Non-wood**
Cycle growth	➢ Long growth cycles [X]	➢ Short cycle growth [√]
Cellulose content	➢ Higher cellulose content [√]	➢ Lower cellulose content depends on the types of non-wood [X]
Lignin content	➢ Contain higher lignin content [X]	➢ Contain lower lignin content [√]
Chemical uses	➢ Use a large volume of chemical during pulping process [X]	➢ Use a small amount of the chemical in pulping process [√]
Time pulping	➢ Need long time for pulping process [X]	➢ Shorten time for pulping process [√]
Cost operation	➢ Expensive due to the limitation resources [X]	➢ Cheaper cost because the abundance resources [√]
Environmental impact	➢ Increase environmental problem such as global warming and soil erosion [X]	➢ Reduce environmental impact which reduce the deforestation problem and improve sustainable forestry [√]

Note: [√] = advantages and [X] = disadvantages

2.4 Non-wood fibre resources

Nowadays, in paper making industry, the environmental problems have brought forward the need for cleaner technology where the new non-wood resources have been introduced to replace traditional raw materials such as wood resources with non-wood resources. The cleaner technology or green technology is applied to achieve increased production with minimum effect especially on the environment and lessen the disposal cost, steadiness risks and resource cost resulting in a declined burden on the natural environment and also increase the profits in pulp and paper-based industries (Sridach, 2010b). The abundance of non-wood fibres in some countries, made them responsible for its use in pulp and paper-based. This is considered as the best way and more profitable for non-wood fibre to be used as alternative fibres in paper-based production.

There is a growing interest in the use of non-wood resources in pulp and paper-based industries. There are many studies about the potentiality of non-wood plant species which are tobacco (Shakhes *et al.*, 2011a), wheat straw (Jiménez *et al.*, 2002a), giant reed (Shatalov and Pereira, 2006), canola straw (Hosseinpour *et al.*, 2010), Tunisian alfa (Marrakchi *et al.*, 2011) and vine (Mansouri *et al.*, 2012) stems as a good fibre resources to replace the wood fibre resources in pulp and paper-based industries. Figure 2.4 represents the percentage of types of non-wood pulp used in paper production.

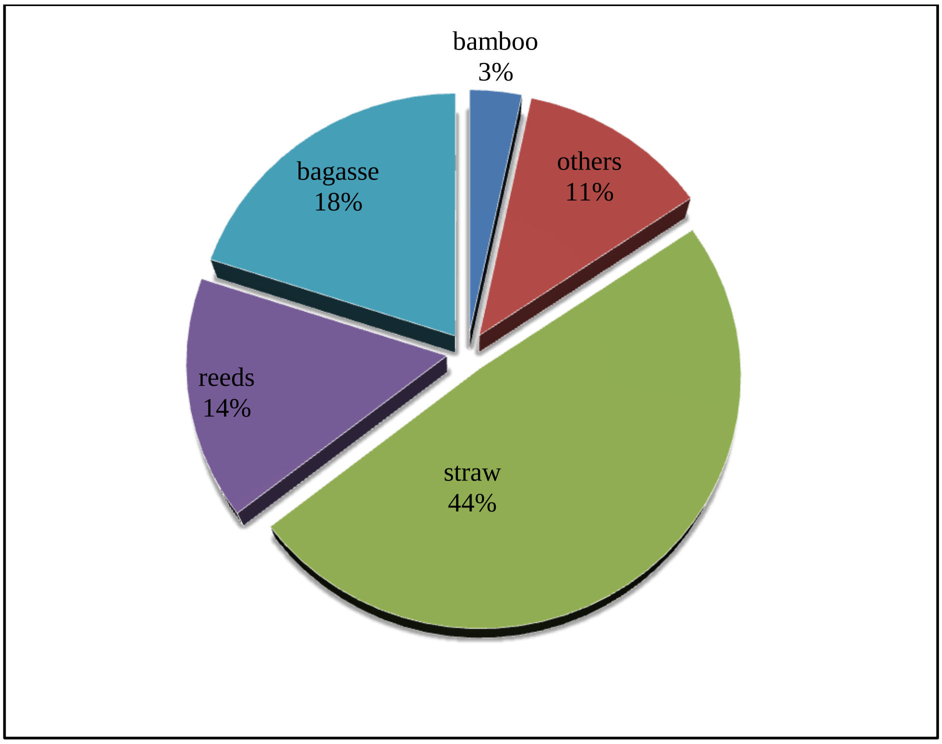

Figure 2.4: Consumption of types of non-wood pulp in paper production (Sridach, 2010b)

In Asia, the production of non-wood resources as pulp for paper production mainly takes place in countries with lack of wood supply especially in China, where it produces around more than two thirds of the non-wood pulp worldwide for paper and paperboard production (Hammett *et al.*, 2001). Bangladesh and Vietnam use non-wood (jute and bamboo respectively) as alternative fibres in pulp and paper – based to replace origin wood fibre and increase their paper production (Bay, 2001 and Jahan *et al.*, 2009).

In addition, Europe and America also use non-wood resources such as agricultural residues (hemp and wheat straw) in pulp and paper-making because it averts the need for disposal, which is currently increasing farming costs and environmental deterioration through pollution, fires and pests (Chandra, 1998). Furthermore, oil palm fibres (WanRosli and Law, 2011), kenaf (Ibrahim *et al.*, 2011) and banana stem fibre (Abd Rahman and Azahari, 2012) are examples of non-wood sources that are being studied for pulp and paper-based industries in Malaysia because of the abundant sources, to decrease disposal into landfill and to prevent deforestation activities.

High market demands as well as the environmental issues because of the large usage of wood supply in pulp and paper production have increased the interest to seek for non-wood plants as substitution fibre which is also environmentally friendly (González-García *et al.,* 2010 and Jiménez *et al.,* 2002a). Therefore, the consumption of non-wood fibre is a better solution for producing pulp and paper to reduce deforestation of rain forests or primeval forests in the world including Malaysia.

In the case of specialty paper production, non-wood plants are the raw materials for production of high-quality specialty papers (Gomihno *et al.,* 2001 and WanRosli *et al.,* 2004). Non-woody plants have given many benefits as can be seen in Table 2.3. Moreover, an additional benefit for these fibre resources is it can give additional income to the farmers for food crop-waste such as straw, bagasse and grasses (Salmela *et al.*, 2008).

Apart from the above reasons, some non-wood plant fibres are in demand for pulp and paper-making due to the special properties that make them better than wood fibre. For example, abaca is an excellent raw material for manufacturing of specialty paper, for its long fibre length and high strength properties such as tear, burst and tensile indices (Peralta, 1996). In addition, sisal can be made into strong products whereas cotton linters are used for premium quality letterhead paper, currency paper, dissolving pulp and other specialty products (Chandra, 1998). Moreover, bagasse and straw are best at contributing excellent formation to papers and can replace hardwood chemical pulps for printing and writing paper (Sridach, 2010b).

Generally, non-wood plant fibres that are used in pulp and paper industries can be broadly divided into three categories based on their availability. These are agricultural residues, natural growing plants (annual plants) and non-wood crops grown primarily for their fibre (Sriadch, 2010b). First, agricultural residues are considered by low raw materials price, moderate quality and the abundance of raw materials after harvesting season such as rice (Navaee-Ardeh *et al.,* 2004), wheat straw (Jiménez *et al*., 2002b), corn stalk (Flandez *et al.*, 2010) and sugarcane bagasse (Hemmasi *et al.,* 2011).

The naturally growing plants which are the type consist of bamboo (*dendrocalamus strictus*), reeds (*Phragmites communis* Trinius), sabai grass

(*Euaiopsis binata*), papyrus (*Cyperus papyrus*) and elephant or napier grass (Madakadze *et al.*, 2010). Fibre from bast fibre, Jute (*Corchorus capsularis*), ramie (*Boehmeria nivea*), leaf fibres, abaca (*Musa textiles*), seed hair fibre, cotton fibre, rags and linters and kenaf (*Hibiscus cannabinus*) are the third category of non-wood fibre that are the most important resources in pulp and paper making (Chandra, 1998).

Table 2.4 shows the summary of properties for various non-wood fibres in pulp and paper-based industries. Agricultural residues have higher cellulose and lower lignin content than annual plants and non-wood crops. These contents generally provide the higher mechanical properties of handsheet. The chemical and physical properties of non-wood fibres also affect their mechanical properties. For example, Elephant grass, in annual plants contains lower lignin and short fibre length that contribute to high strength property.

Table 2.4: Summary of properties for non-wood plants

Categories	Raw materials	Chemical properties			Physical properties		Mechanical properties		
		Cellulose, w/w %	Hemicellulose, w/w %	Lignin, w/w %	Fibre length, mm	Fibre diameter, µm	Tensile index, mN/g	Tear index, mN.m²/g	Burst index, kPa.m²/g
Agricultural residue	Banana stem (*Musa paradisica*)[a]	59.18	17.50	18.21	1.55	22.00	47.76	9.10	4.51
	Rice straw[b,c]	41.20	19.50	21.90	1.41	8.00	26.11	0.31	1.20
	Sugarcane bagasse[d]	42.34	28.60	21.70	1.51	21.40	58.00	5.80	4.20
	Wheat straw[b,e]	38.20	36.30	15.30	0.74	23.02	76.70	4.11	3.74
Annual plant	Bamboo[f]	43.00	39.00	31.00	2.70	14.00	n.a	18.10	4.90
	Elephant grass[g]	45.60	n.a	17.70	0.75	15.14	93.25	4.40	5.85
	Switch grass[g]	41.20	n.a	23.89	0.76	13.89	75.98	5.60	4.90
Non-wood crops	Kenaf bast[h]	55.50	17.70	12.50	2.90	28.16	2.09	11.84	n.a
	Palmyra plant fruit[i]	37.01	31.51	18.54	1.07	n.a	13.80	1.12	n.a
	Date palm rachis[j]	45.00	29.80	27.20	0.89	22.30	n.a	4.40	1.32
	Date palm leaves[j]	30.30	n.a	31.20	n.a	n.a	28.30	8.40	1.40

n.a: non-available, a: Goswami *et al.* (2008), b:Enayati *et al.* (2009), c: Sridach, (2010b), d: Agnihotri *et al.* (2010), e: Berrocal *et al.* (2004), f: Sarwar *et al.* (2009), g: Madakadze *et al.* (2010), h: Udohitinah & Oluwadare, (2011), i: Sridach, (2010a), j: Khiari *et al. (*2010)

Therefore, with the best intention of solving disposable issues of agricultural in Malaysia and finding the best alternative for waste non-wood pulp, this study is focused on cassava peel, cocoa pod husk (agricultural residues), cogon grass (natural growing plants) and oil palm leaf (non-wood crops grown primary) due to their abundance and less utilisation of these waste materials. In addition, the concepts of “from waste to wealth” and “recyclable material” are now important in Malaysia in order to build a sustainable and sound material-cycle society through the effective use of these waste resources.

2.4.1 Cassava peel

Cassava (*Manihot esculenta Crantz*), a member of the *Euphorbiaceae* is a perennial shrub which origin is from the Amazon basin. Cassava cultivation has now spread throughout the humid tropics from Latin America to Africa and Asia (Buschmann *et al.*, 2002). Cassava is a staple food in the world especially in Tropical Africa, Nigeria and Central and South America (Ezekiel *et al.*, 2010). In addition, cassava is also fast becoming a marvel crop due to its potential usage in several agro and agro-allied industries. Nigeria is the world’s leading producer of cassava followed by other producer countries such as Brazil, Zaire, Thailand, Indonesia, China, Malaysia, Malawi, Togo and Tanzania (Olukunie *et al.*, 2010).

Cassava peel (Figure 2.5) is an agricultural waste from the food processing industry (Adesehinwa *et al.*, 2011). The thickness of cassava peel varies between 1 to 4 mm and may account for 10 to 13% of the root total dry matter (Ezekiel *et al.*, 2010 and Olanbiwoninu and Odunfa, 2012). Malaysia has large cassava plantations (2,769 ha in 2010) that yield about 68,508 tons of roots during harvesting season (MOA, 2011). Therefore, the waste of cassava peels produced in 2010 is estimated at 6,900 to 8,900 tons. The explosive development of cassava production in this country has generated massive amounts of cassava peels as waste materials that are dumped in landfills and allowed to rot in the area, creating a great environmental problems in the long run especially in health hazard (Oboh, 2006).

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